Search for fractionally charged particles in sea water, ocean sediment deposits, and volcanic lava

D. D. Ogorodnikov, I. M. Samoĭlov, and A. M. Solntsev

I. V. Kurchatov Atomic Energy Institute
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The particles sought are extracted from matter by thermal desorption, accumulated by application of an electric field, and then identified. Upper limits for the concentration of quarks determined from the measurements for various materials studied lie in the range 5×10^{-28}–6×10^{-25} quarks/nucleon.

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According to the well known hypothesis of Gell-Mann and Zweig, fundamental elementary particles with fractional electric charge—quarks—may exist.[1,2] Many years of attempts to observe these particles by different methods have so far failed to give positive results.[3,4] However, the possibilities of searching for quarks in matter[3–8] have not been exhausted to the same degree as in experiments in contemporary accelerators and in cosmic rays.[1)] The lowest experimental limits for quark concentration were obtained in Refs. 7 and 8 for solid rocks and sea water (10^{-23} and 10^{-24} quarks/nucleon, respectively). However, the experiment of Cook *et al.*[7] was sensitive only to quarks with mass up to 5–10 GeV, while in the work of Chupka *et al.*[8] for water a limitation can be imposed by the low temperature of desorption of the particles containing quarks from the salt left after evaporation of the water (400 °C).

The selection of a method of search is limited by the difficulty in predicting the physical and chemical parameters of the quark-ions sought, which are formed on contact of quarks (or polyquarks) with ordinary matter. Therefore from the point of view of complete realization of the assumptions on which the search is based, materials containing the largest number of possible forms of quark-ions are preferred. This important requirement is satisfied by the ocean.

We undertook to search for fractionally charged particles in sea water, ocean sediment deposits: iron-manganese concretions, finely dispersed clays, radiolarian ooze with foraminifera,[10] and volcanic lava. A comprehensive study of the ocean is necessary as the result of the possible depletion of the water in quark-ions (up to $\sim10^7$ times) and the preferential accumulation of them by the deposits indicated.[5,6,9,10] We note that clays, as the product of erosion of mountain rocks, should contain also quarks of continental origin (from cosmic rays), the interest here being primarily in quark-ions of the easily removed (by heating up to 400–600 °C) water of crystallization which occurs in all the main mineral components of clays. Volcanic lava was considered as a source of residual quarks. To extract the desired particles we used desorption on heating the materials to 700–800 °C. Estimates show that with this method we can count on efficient extraction of particles not only from the surface of micropores (in a time of $\sim10^3$–10^5 sec), but also from an appreciable fraction of the volume, since all of the materials investigated by us are finally dispersed and have a porous structure. The method used was similar to that of Chupka *et al.*[8] Quark-ions extracted from matter were first accumulated on electrodes of an electrical filter. Then they were transferred to a special collector which subsequently served as an emitter of quark-ions in the source of the apparatus in which the particles were identified. The ions leaving the source were accelerated by a voltage of 20–25 kV and focused onto a detector consisting of an open electron multiplier. Analysis of the ions can be accomplished either by a magnetic mass spectrometer or by a special technique of recording desorption curves of ions with use of an electrical gate with which the source was supplied (see below). The search was carried out on the theory that quarks with charges of both signs were present.

ACCUMULATOR OF QUARK-IONS

A diagram of the accumulating apparatus is shown in Fig. 1. The solid materials were loaded into a crucible 1 in amounts up to 0.5 kg in layers of thickness 0.5–1 cm. Each sample of water consisted of 5 liters. On heating of the crucible (up to 700–800 °C) the volatile

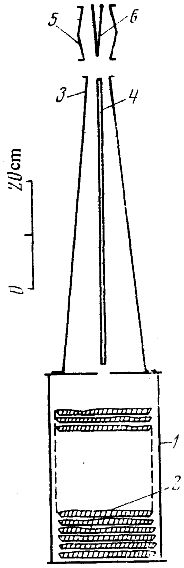

FIG. 1. Diagram of accumulator apparatus: 1—crucible; 2—plates with material; 3, 4—filter electrodes (3—ground, 4—high voltage, tube diameter 1 cm); 5, 6—collector trap electrodes; the collector 6 is made in the form of a V-shaped platinum filament of diameter 0.3 mm. The parts 1–4 are made of stainless steel.

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fractions and vapors of the material passed through a pipe consisting of an electrical filter formed by electrodes 3 and 4 and evacuated by a pump; here the pressure of the gas (or vapor) in the accumulative volume was in the range ~ 0.1–10 Torr (depending on the volatility of the material). Quark-ions were picked up by the electrodes 3 and 4 from the flow as the result of a potential $V_f = \pm(100-120)$ V applied to electrode 4. At the end of the accumulation stage they were transferred (on removal of V_f) to a collector 6 introduced into the accumulative volume (through a vacuum lock) immediately before beginning the transfer. For appropriate potentials of the collector ($V_c = \pm 150$ V) and of the screening electrode 5 ($V_s \approx 0.3V_c$) the collector caught charged particles of one sign, and particles with the opposite charge were retained in the filter volume. At the end of the transfer the potential V_f was again applied, the collector was removed, and a new collector for quark-ions of the other sign was installed in its place. In the transfers of particles to the crucible we used a small flow of nitrogen, which also passed through the filter.

The question arises of the efficiency of operation of the filter during the long period of accumulation (~ 10^4–10^5 sec). Evaporating on the average after a time τ, the quark-ions on their being returned by the field E to the initial electrode do not fall on their previous locations, as a consequence of diffusion by scattering in the gas and entrainment by the directional flow of vaporized material. In boundary-layer migrations of this type a quark-ion with charge q is removed from the electrode by a distance $\Delta \sim D/\mu E = T_f/qE$ and spends in the gas a time $\theta \sim 2\Delta^2/D$, where μ and D are the mobility and diffusion coefficient of the particles and T_f is the temperature of the electrodes and the gas. If appreciable surface ionization of quark-ions on the electrodes or charge exchange of the ions in the gas is not excluded, then in both cases transfers of particles between electrodes under the influence of the field E become possible; the time spent by them in the gas rises rapidly in comparison with θ, and accordingly their drift, diffusion, and migration over the filter increases. It is evident that quark-ions will be held in the filter at least for the time t_f until they reach its open exit end, where loss of particles is not excluded. If we take into account respectively only particle drift or only scattering, t_f can be represented in the form

$$t_f \sim (L/s) K_\alpha \tau + \delta t\ , \quad t_{fD} \sim (L/s_D)^2 K_\alpha \tau + \delta t\ ,$$

where δt is the time spent by the quark-ions in the gas, K_α is a coefficient taking into account the effect of surface ionization or charge exchange, L is the length of the filter, and s and s_D^2 are respectively the drift of a quark-ion in the vapor flow and the mean square displacement as the result of diffusion in one cycle of evaporation and return to the initial electrode. Expressions for the quantities δt, K_α, s, and s_D are given in Table I; they refer to operation with a quark-ion mean free path $\lambda < \Delta$. In the general case $t_f = K\tau$ if we neglect the contribution due to δt. In the boundary-layer migrations the diffusion mechanism is important—it is universal and acts independently of the value of N. With our filter parameters $K = K_D \sim 5 \times 10^8$ (retainment on electrode 4, Fig. 1, at $T_f = 300$ K) while the contribution due to drift of quark-ions is comparable with it only for very large values of $N \sim 1$ g/sec. The limitations due to diffusion are insignificant—for a period of 10^4–10^5 sec quark-ions are reliably retained with $\tau \geq 10^{-5}$–10^{-4} sec.

For migrations due to surface ionization and charge exchange, $K = K_D \sim 3 \times 10^4 K_\alpha$, and the contribution due to drift is already comparable for $N \sim 10^{-3}$ g/sec, i.e., under these conditions the efficiency of the filter is essentially limited by the drift. Here for moderate working values $N \sim 10^{-2}$ g/sec we have $s \sim 10^{-2}$ cm and $K = K_N \sim 5 \times 10^3 K_\alpha$. Under extreme conditions where $K_\alpha \sim 1$ and t_f is minimal, the filter is obviously inefficient (particles with $\tau \lesssim 1$–10 sec cannot be retained). However, in the first case the danger is high only for quark-ions with ionization potentials V_q comparable with the work function φ of the surface, since already for $|V_q - \varphi| \gtrsim 0.3$ V we have $K_\alpha \sim 10^5$ and $K \sim 10^9$. Charge exchange affects all particles with $V_q < \varphi$ and this is more dangerous. The extreme condition corresponding to $V_q = \varphi$ in this case is the closeness of the gas pressure p to some pressure p_m which depends on the effective charge-exchange cross section $\sigma_{eff} = (\sum_i n_i \sigma_i)/\sum_i n_i$. Here in the worst case we estimate $K_\alpha \sim 1$, and $p_m \sim (10^{-17}/\sigma_{eff})^{1/2}$. However, for charge exchange of quark-ions at thermal velocities it is necessary that there be present in the gas particles with ionization potentials $V_i \lesssim \varphi$.

In our case the probability of realization of unfavorable processes of ionization or charge exchange simultaneously for several forms of quark-ions should be small even when we consider the uncertainty in the state of the actual electrode surfaces. However, this does not remove the rigid requirements on filter efficiency. To increase it, the electrodes were water cooled during the accumulation, for $t_f \sim \tau \sim \exp(Q/T_f)$, where Q is the binding energy with the surface.[12] In addition, the transfer was carried out one stage at a time with filter-electrode temperatures T_f equal to 30, 200, and 500 °C (onto new collectors for each T_f), which was done to decrease the contamination of the collectors and to reduce the possibility of loss of particles with relatively small Q from the filter during the heating of the electrodes before transfer of the particles to the collectors.

MEASUREMENT TECHNIQUE AND RESULTS

To reduce the flux of background ions in the apparatus where particle identification was carried out, we used indium gaskets and evacuation by sorption pumps.[11] Figure 2 shows a diagram of the source apparatus, which served as an emitter of quark-ions in the mass spectrometer. The collectors were placed on the inner wall of the crucible 1. The needle and crucible were maintained with respect to the enclosure 6 at constant (keep-alive) potentials of the same sign (44 and 22 V, respectively), which played the role of an electrical gate. They were removed by means of an electronic circuit only immediately at the time of detection; their sign was such that the quark-ions were retained by the

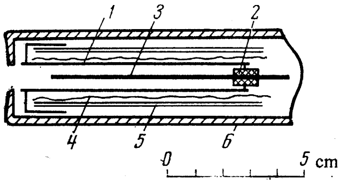

FIG. 2. Diagram of the source apparatus: 1—source crucible (tube with wall thickness ~ 0.5 mm), 2—ceramic insulator, 3—needle 1 mm in diameter, 4—heater, 5—heat shields, 6—enclosure. Parts 1 and 3 are made of stainless steel and are joined by an insulating vacuum seal 2.

needle. The desired particles were desorbed by heating the source crucible to 400–600 °C.

In preliminary experiments we observed spurious effects similar to those observed in Ref. 8: On heating the source, instead of falling desorption curves we observed either the absence of a drop or a burst of intense counting after the cycle of turning on and turning off the electrical gate and reversal of the sign of the field accelerating the ions. Mass-spectrometric monitoring showed that these effects were related to contamination of the collectors by vapors of the material in the accumulator. It was observed in addition that background negative ions ($\sim 10^3$–10^4 ions) appeared from a "clean" source crucible after its contact with the atmosphere. To remove these effects we introduced a special measurement technique (see below) and an intermediate transfer of quark-ions from the accumulator collectors (A) to "clean" collectors similar to them (B). This transfer was carried out in a nitrogen atmosphere at a pressure of ~ 1 Torr with the filament A heated to ~ 800 °C and collector B at a temperature 20 °C.

In most of the main experiments the flux of charged particles from the source was below the sensitivity of the mass spectrometer. In view of this, the detection at the mass-spectrometer input was carried out according to the following program. With the gate closed the temperature of the source crucible T_{sc} was raised in steps of ~ 100 °C. The source was soaked at each step T_{sc} for a period $t_s = 5$–10 min, and the gate was opened for time intervals t_1 and t_3 separated by a closed interval t_2. In most of the experiments $t_{1,2,3} = t_i = 0.1$ sec;

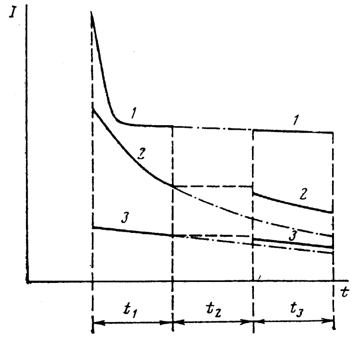

FIG. 3. Schematic representation of the expected desorption curve intervals for the desired particles in the presence of background ions. Curves 1, 2 and 3 are respectively for $\tau \ll t_i$, $\tau \sim t_i$, and $\tau \gg t_i$.

in the last steps of T_{sc}, the values of t_i were 0.1, 1.0, and 10 sec. During t_1 and t_3 we measured the change in the charged-particle counting rate and their total numbers N_1 and N_3. If the desired particles are present we can expect the intervals in the desorption curves to have the features shown schematically in Fig. 3. The desired effect can be characterized by the difference $\Delta N = N_1 - N_3$. The contribution to ΔN from background particles is small, since at the time of the measurements there may remain in the source ordinary (background) particles with adsorption times $\tau_f \sim t_s \gg t_i$ whose changes in intensity are small ($\Delta N \sim N_{1f} 2t_i/\tau_f$). The sensitivity of this technique is determined mainly by fluctuations of the numbers N_1 and N_3. Step-by-step detection in the steps T_{sc} permitted most background ions to be avoided without greatly reducing the lifetimes of the desired particles in the source. We note that the observation method used is limited in the sense that the existence of the effect is a necessary but not a sufficient condition for identification of the particles as fractionally charged.

In the course of the measurements in identification both of particles with charge $q < 0$ (in the range $T_{sc(-)} = 25$–470 °C) and of particles with charge $q > 0$ ($T_{sc(+)} = 25$–280 °C), in none of the experiments did we observe effects suggesting the particles sought, and as a rule the counting rate did not exceed the intrinsic background of the heated source and amounted to $I_{(-)} \sim I_{(+)} \sim 1$–$10$ counts/sec (at $T_{sc} = 25$ °C, $I_{(-)} \sim I_{(+)} < 1$ count/sec). In the higher stages of $T_{sc(-)} = 570°$ and $T_{sc(+)} = 370$ °C the counting rate rose, but in most cases within the fluctuations of $N_1 = N_3$ ($I_{(-)} \lesssim 10$ counts/sec, $I_{(+)} \lesssim 10^4$ counts/sec—the background of ions from the alkali metal impurities of the source material). The only exception was one series of experiments with lava, in which in all four samples we recorded ΔN from 20 to 100 par-

TABLE I.

Movement due to drift	Movement due to diffusion
Boundary-layer migrations	
$s \sim 2v_s\Delta^3/RD$	$s_D \sim 2\Delta$
$\delta t \sim LR/v_s\Delta$	$\delta t_D \sim L^2/2D$
$K_\alpha = 1$	$K_\alpha = 1$
With inclusion of surface-ionization effect	
$s = v_s(R_2 - R_1)\mu\bar{E}$	$s_D = [4T_f(R_2 - R_1)/q\bar{E}]^{1/2}$
$\delta t = L/v_s$	$\delta t_D \sim L^2/2D$
$K_\alpha \sim 2 + \alpha + \alpha^{-1}$	
With inclusion of charge-exchange effect	
$s = v_s(R_2 - R_1)\mu\bar{E}$	$s_D = [4T_f(R_2 - R_1)/q\bar{E}]^{1/2}$
$K_\alpha \sim \frac{1}{2}\ln[\ln(R_2/R_1)qV_f/4T_f]$	

Note. The following designations are used: $v_s = N/n\pi(R_2^2 - R_1^2)$ is the average directed velocity of particles of the vapor, whose flux is N particles/sec with a density n cm^{-3}; $\alpha \sim \exp[-(Vq - \varphi)/T_f]$ is the coefficient of positive surface ionization of quark-ions; $\bar{E}$ is the field at the average radius $\bar{R} = (R_1 + R_2)/2$. The effect of charge exchange was estimated for the extreme conditions, for which t_f was minimal; R_1 and R_2 are respectively the radius of the inner electrode 4 of the filter (Fig. 1) and of the conical tube 3 in its central portion.

TABLE II.

Materials studied	Upper limits of quark concentration (quarks/nucleon)			
	In detection of negative particles		In detection of positive particles	
	$T_{sc}=25\,°C$	$T_{sc}=25-470\,°C$	$T_{sc}=25\,°C$	$T_{sc}=25-280\,°C$
Sea water (20 liters)	$5\cdot10^{-28}$	$3\cdot10^{-27}$	$5\cdot10^{-28}$	$4\cdot10^{-27}$
Finely dispersed clays (2 kg)	$5\cdot10^{-27}$	$5\cdot10^{-26}$	$5\cdot10^{-27}$	$4\cdot10^{-26}$
Radiolarian ooze (2 kg)	$5\cdot10^{-27}$	$3\cdot10^{-26}$	$5\cdot10^{-27}$	$4\cdot10^{-26}$
Concretions (1 kg)	$7\cdot10^{-27}$	$6\cdot10^{-25}$	$7\cdot10^{-27}$	$3\cdot10^{-25}$
Volcanic lava (2 kg)	$5\cdot10^{-27}$	$2\cdot10^{-26}$	$5\cdot10^{-27}$	$1\cdot10^{-25}$ $(2\cdot10^{-25})$*

*The effect for $T_{sc}=370\,°C$ from the differences N_1 and N_3; see the text.

ticles (only for $q>0$), and in two of the four experiments $\Delta N \gtrsim 3\sqrt{(N_1+N_3)/2}$. This was observed only at $T_{sc(+)} = 370\,°C$ and $t_i = 0.1$ sec and only for those collectors to which quark-ions with $q>0$ were transferred at the very high temperature $T_f = 500\,°C$. The set of data presented does not permit explanation of these weak effects in a simple way—by ordinary intensity fluctuations. Unfortunately, with such small ΔN values it is difficult to establish the nature of the particles responsible for the unusual behavior, even with the aid of the mass spectrometer (in our apparatus in the mass range searched up to ~ 200 amu at least 10^4 particles are required); see also Ref. 8. We can only assume that these effects can be produced by dielectric films brought into the source (with the collectors), which are capable of accumulating ions during the soaking time t_s, or by the presence in the lava of particles with ionization potentials $\lesssim 4$ V. Association of these effects with the existence of quarks would be extremely optimistic as long as there is any possibility of interpreting them as the unlikely behavior of ordinary particles.

Thus, the total result of our search must be considered negative. In Table II we have given estimates of the upper limits of quark concentrations in the materials studied by us, on the basis of the total number of particles recorded in all experiments (but not based on the differences ΔN), on the assumption that they are the particles sought. The upper limits of quark concentration at high T_{sc} are one to three orders of magnitude below the upper limits of concentrations obtained for the corresponding materials in Refs. 7, 8, and 13. It is important that our results were obtained for both signs of charge of the quark-ions and for temperatures to which the materials were heated almost a factor of two higher than in Ref. 8, which substantially increases the reliability of the search in view of the exponential dependence of the desorption time on temperature.

The accumulation of negative results in searches for quarks by different methods, in spite of their limitations, significantly reduces the probability of existence of quarks in the free state. The most rigid limitations of this probability follow from the search for quarks in matter, and this refers also to polyquarks with charges 4/3, 5/3, 7/3, and so forth.

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1) For simplicity and brevity, the discussion will be carried out on the assumption that quarks exist.

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